Adiabatic mechanism of the multiply charged ion production by a laser field through ATI states of an atom

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Abstract

ATI can be followed by an inelastic collision of the ionized electron with the parent atomic particle resulting in an excitation of the ion. It may be a continuum state excitation producing the doubly charged ion or a discrete state which also enhances the doubly charged ion production. Absorption of a few quanta above the atomic threshold is sufficient to make this mechanism work. As a result the two-electron processes can take place even in moderate fields. The example of two-electron excitations of He atoms in a 780 nm laser field with intensity above 10^{14}W/cm^2 is discussed.

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The purpose of this paper is to demonstrate that ATI plays a very important role in multiply charged ion formation in a laser field. The basic physical idea is as follows. Suppose that single electron ionization takes place. After that the ionized electron can undergo an inelastic collision with the parent atomic particle. The electron impact can result in the excitation of the ion into a discrete or continuum state because the ionized electron, strongly interacting with the laser field absorbs and accumulates the high energy necessary for the ion excitation. This energy manifests itself in the form of the electron wiggling in the laser field as well as in the translational motion of the electron. Consider the classical momentum of the electron. The laser field results in the oscillation of the momentum $\mathbf{p}(t)$, $\mathbf{p}(t) = \mathbf{k} + \mathbf{f}\sin\omega t$. Here the first constant term $\mathbf{k}$ describes the translational motion and the second term, $\mathbf{f}\sin\omega t$, where $\mathbf{f} = \mathbf{F}/\omega$, describes the wiggling of the electron in the laser field which is considered to be linearly polarized: $\mathbf{F}(t) = \mathbf{F}\cos\omega t$. As a result the "energy" $E_{\mathbf{k}}(t)$ of the electron is also time-dependent, it behaves like

$$E_{\mathbf{k}}(t) = \frac{1}{2}\left(\mathbf{k} + \mathbf{f}\sin\omega t\right)^2 . \tag{1}$$

For a given level in the ATI spectrum the translational momentum satisfies the energy conservation law $k^2/2 + f^2/4 + E^{(A^+)} = n_1\omega + E^{(A)}$, where $E^{(A)}$ and $E^{(A^+)}$ are the energies of the atomic and ionic ground states, n_1 is the number of absorbed quanta during the single electron ionization. Obviously for higher energy levels in the ATI spectrum, *i.e.* for larger n_1, the translational momentum is greater. (For description of ATI see Refs. [1,2] and references in the review [3].) We will show below that double ionization caused by inelastic scattering of the firstly ionized electron on the parent atomic particle is possible if the energy necessary for removal of the second electron satisfies a condition

$$E^{(A^{++})} - E^{(A^+)} \leq \frac{1}{2}\left(k + f\right)^2 \ , \tag{2}$$

where $E^{(A^+)}$ and $E^{(A^{++})}$ are the ground states of the single and doubly charged ions. Eq.(2) has a clear physical meaning. Consider a given energy level in the ATI spectrum. Then the right-hand side of Eq.(2) is equal to the maximum of $E_{\mathbf{k}}(t)$ (1), $\max[E_{\mathbf{k}}(t)] = (1/2)(k+f)^2$, which is achieved when $\mathbf{k}/k = \pm\mathbf{F}/F$ and $\omega t = \pm\pi/2$. Inequality (2) simply states that the maximal energy of the photoelectron, which is possible for a given n_1, must exceed the ionization energy. If the field is strong enough, *i.e.* $f^2/2 \geq E^{(A^{++})} - E^{(A^+)}$, then according to Eq.(2) it is sufficient to consider the lowest level in the ATI spectrum for which $k \approx 0$. For

weaker fields the translational momentum becomes vital and therefore ATI comes into play. For a sufficiently high level in the ATI spectrum the translational momentum becomes high enough to satisfy the inequality in Eq.(2) permitting the mechanism to work. Note that there is no sharp threshold for the field intensity, below which the mechanism fails. It always works, but the weaker the field, the higher the necessary level in the ATI spectrum.

It is important that according to Eq.(2) the necessary level in the ATI spectrum can be well below the energy necessary for removal of the second electron. The above-threshold energy $E_{\rm ati}$ of the level in the ATI spectrum $E_{\rm ati} = k^2/2 + f^2/4$ is always lower than the maximal kinetic energy of the photoelectron on this level, $k^2/2 + f^2/4 < \max[E_{\mathbf{k}}(t)] = (1/2)(k + f)^2$. The stronger the field, the more pronounced the difference between $E_{\rm ati}$ and $\max[E_{\mathbf{k}}(t)]$. As a result $\max[E_{\mathbf{k}}(t)]$ can exceed the ion excitation energy while $E_{\rm ati}$ can be well below it, $E_{\rm ati} < E_{\rm exc} < \max[E_{\mathbf{k}}(t)]$. This means that absorption of a small number of quanta above the atomic threshold can put the considered mechanism in action, resulting in the population of the doubly charged ion state.

The firstly ionized electron, which plays a crucial role in the considered mechanism, can be looked at as a kind of antenna. It accumulates the energy from the field and transfers it to the parent atomic particle. This idea was first considered in Ref. [4]. It was shown in this work that the scattering of the ionized electron on the parent ion can result in the ion excitation or double ionization. It can also increase the energy of the firstly ionized electron populating the very high levels in the ATI spectrum. These results were based on the analytical calculation of the amplitude of the double ionization process. The calculations are vital to justify the physical idea of rescattering. Without them one can doubt that a collision of the ionized electron with the ion is possible at all because during single electron ionization the ionized electron can go far outside the atom and never return. The calculations described in Ref. [4] and presented in detail in Ref. [5] show that there is no danger of this kind, the collision is very probable. The physical idea of rescattering of the ionized electron on the parent atomic particle was also discussed in the recent paper Ref. [6]. In this work a model approach was developed. Recent experiments with two color lasers on ATI spectra Ref. [7] support the rescattering mechanism. In Ref. [8] the results of precise measurements of double ionization of Helium are presented and compared with the rescattering mechanism. A conclusion of this work is that the experimentally observed yield of doubly charged ions is

higher than predicted by the rescattering mechanism. The recent work Ref. [9] also indicates that the production of doubly charged due to the rescattering mechanism is smaller than the one observed experimentally. Note, however, that this discrepancy is not strong, one or two orders of magnitude. At the same time there are uncertainties in the calculations. They come from uncertainties in the cross sections of He^+ ionization and excitation in the laser field and uncertainties connected with the classical models developed in Refs. [6] and [9]. Therefore, the rescattering mechanism qualitatively agrees with the recent experiments Ref. [8] as well as with earlier experiments on noble gases Refs. [12–18].

Let us demonstrate the validity of inequality (2). Consider an atom placed in the laser field whose frequency is much lower than the excitation energy of the atom. The fact that the frequency is low permits one to consider the process as adiabatic. This is the basis of the Keldysh solution of the single electron ionization problem, Refs. [10], [11]. Consider what happens with the system after the single electron ionization takes place. The adiabaticity allows one to describe the strong influence of the laser field on the electron system using the time-dependent "energy" of the electron system. Following the tradition of the theory of adiabatic processes we will call this "energy" an electron *term*. To simplify consideration we will neglect the static Coulomb interaction between the ionized electron and the ion. This approach was first used in Ref. [10]. It is supposed to give a reasonable description of atomic ionization and must be even better for electron detachment from negative ions. The system under consideration is the single-charged ion and the ionized electron. We will neglect the influence of the laser field on the energy $E^{(A^+)}$ of the ion ground state because it is suppressed due to a high ionization potential of the ion. In contrast, the energy of the ionized electron exhibits strong variation. Eq.(1) describing this variation was evaluated from the classical point of view, but it remains valid for the quantum description as well. This follows from the Volkov solution, Ref. [19], for the electron wave-function in a laser field: $\psi_{\mathbf{k}}(\mathbf{r},t) = \exp\{i[(\mathbf{k}+\mathbf{f}\sin\omega t)\mathbf{r} - \int_0^t E_{\mathbf{k}}(\tau)d\tau]\}$, where $E_{\mathbf{k}}(t)$ in the exponent describes the term variation given in Eq.(1). Thus the term $E^{(\mathrm{e+A^+})}(t)$ of the system consisting of the single-charged ion and the photoelectron is

$$E^{(\mathrm{e+A^+})}(t) = E^{(A^+)} + E_{\mathbf{k}}(t) \ . \tag{3}$$

Consider now the final state of the reaction where there are the doubly charged ion and two electrons in the continuum. Let us neglect the static Coulomb field of the ion as well as

the Coulomb repulsion between the ionized electrons. This approximation is certainly more questionable than the similar one for the single-electron ionization. Nevertheless, one can suppose it to be reasonable for double electron detachment from negative ions. It is argued below that the case of atomic ionization can also be approached using this approximation. Considering the energy of the doubly charged ion as a constant $E^{(\mathrm{A}^{++})}$ and taking into account that the energies of both ionized electrons oscillate in accordance with Eq.(1) we find the final state term of the system consisting of the doubly charged ion and the two ionized electrons

$$E^{(\mathrm{e}_1+\mathrm{e}_2+\mathrm{A}^{++})}(t) = E^{(A^{++})} + E_{\mathbf{k}_1}(t) + E_{\mathbf{k}_2}(t) \ . \tag{4}$$

Here $\mathbf{k}_1,\ \mathbf{k}_2$ are the translational momenta of the two electrons in the continuum. They depend upon the total number n of absorbed quanta $[k_1^2+f^2/4]+[k_2^2+f^2/4]+E^{(A^{++})} = n\omega+E^{(A)}$, where $E^{(A)}$ is the atomic energy and $n\omega$ exceeds the ionization potential, $n\omega > E^{(A^{++})} - E^{(A)} + f^2/2$.

According to the adiabatic theory a transition from the term of the system "electron + single charged ion" to the term "two electrons + doubly charged ion" can take place with high probability if there is a crossing of the two terms for some real moment of time t:

$$E^{(\mathrm{e}+\mathrm{A}^{+})}(t) = E^{(\mathrm{e}_1+\mathrm{e}_2+\mathrm{A}^{++})}(t) \ . \tag{5}$$

We are interested to find this crossing when the firstly ionized electron occupies some low-lying level in the ATI spectrum because population of lower levels is the most probable. The behavior of the two terms in Eq.(5) is illustrated in Fig.1 for different levels in the ATI spectrum. It is important to consider the final state term of the doubly charged ion given in Eq.(4) when both ionized electrons occupy levels in the ATI spectrum, above the ionization potential of the doubly charged ion. Then this term can possess minima at the moments of time when the term of the single-charged ion Eq.(3) possesses maxima. The example presented in Fig.1 illustrates that the desirable crossing of the two terms can be obtained if a few quanta are absorbed above the atomic threshold, see the crossing of the term **b** with the terms **d** and **e**.

It is easy to verify that the crossing of terms in Eq.(5) is possible if inequality (2) is fulfilled. Therefore, if (2) is valid then the transition from one term to the other is allowed from the adiabatic point of view. In order to prove that the transition really takes place it is necessary to consider a potential having matrix elements mixing the two terms. The role of such a potential is played by the interaction between the ionized electron and the single-charged ion.

It certainly has the necessary matrix elements: they describe the ionization of the second electron caused by the inelastic scattering of the electron on the ion. Having the crossing of the two terms and the mixing potential one can calculate the probability of the transition from one term to the other using perturbation theory. Technically this is a complicated problem because the electron in the system $e + A^+$ belongs to the continuum spectrum. Nevertheless the qualitative final answer to this problem is simple Ref. [5]. The probability $W^{(2e)}$ of the double ionization is $W^{(2e)} \approx W^{(e)}\sigma_{\rm in}/R^2$. Here $W^{(e)}$ is the probability of the population of the lowest level in the ATI spectrum of the single-charged ion which satisfies Eq.(2), σ is the inelastic cross section of the electron impact on the single-charged ion in the laser field. The quantity

$$R = \frac{F}{\omega^2}\{[(\beta+1)^2+\gamma^2][(\beta-1)^2+\gamma^2]\}^{1/2}, \tag{6}$$

is a result of calculations. It can be considered as an effective radius of the region in which the firstly ionized electron is localized. It is proportional to the magnitude F/ω^2 of the electron wiggling in the laser field and depends on the Keldysh adiabatic parameter $\gamma = [2(E^{(A^+)} - E^{(A)})]^{1/2}/f$, and the parameter $\beta = k/f$ describing the considered ATI state of the single-charged ion.

The considered inelastic collision of the ionized electron with the ion takes place when the absolute value of the electron momentum is maximal. Therefore, the neglected influence of the static Coulomb ion field on this electron should not be strong. The final-state electrons appear with the small momenta, and one can expect that the neglected static Coulomb field of the ion can only increase the probability of this event. Note that we also neglected the Coulomb repulsion between the final-state electrons. This was possible because the momenta of the two electrons in the final state can be different, as illustrated in Fig.1 by curves **e**,**d**. These arguments are certainly not decisive and the role of the Coulomb interaction in the final state should be studied in more detail. We will not do it in this work, but consider instead a related problem, in which the final-state Coulomb interaction is less important while the final answer for the problem is similar to the one considered above.

Consider the single electron ionization with excitation of the ion into a discreet state. In this case we have the excited single-charged ion in the final state of the reaction. This excited state can be substantially influenced by the laser field. In the simplest case, when the second order of perturbation theory is valid, the shift of the excited level caused by the field is

$\Delta E^{(\mathrm{A}^{+*})} = -\alpha(\omega)F^2/2$, where $\alpha(\omega)$ is the dynamical polarizability of the considered excited level which is assumed positive, $\alpha(\omega) > 0$. In this approximation one finds the term of the system consisting of the ionized electron and the excited ion

$$E^{(\mathrm{e}+\mathrm{A}^{+*})}(t) = E^{(\mathrm{A}^{+*})} - \frac{1}{2}\alpha(\omega)F^2\cos^2\omega t + E_{\mathbf{k}}(t) \ , \tag{7}$$

where $E^{(\mathrm{A}^{+*})}$ is the non-shifted position of the excited ion level. Now we can consider the transition from the term describing the intermediate state $e + A^+$ to the term describing the final state $e + A^{(+*)}$. This transition is possible if equation $E^{(\mathrm{e}+\mathrm{A}^{+})}(t) = E^{(\mathrm{e}+\mathrm{A}^{+*})}(t)$ is resolved for a real moment of time t. Using Eqs.(3),(7) we find that the solution exists if

$$E^{(\mathrm{A}^{+*})} - E^{(\mathrm{A}^{+})} \leq \frac{1}{2}\left(k + f\right)^2 \ . \tag{8}$$

We see that the ion excitation is allowed if the necessary energy of the excitation is lower than the maximal energy of the photoelectron on the considered level of the ATI spectrum. The resemblance between Eqs.(8) and (2) is obvious.

Notice that there is no sharp energy cut-off for the considered mechanism. Really, it is known starting from Ref. [10] that there is the possibility to populate the ATI levels due to direct absorption of additional, above-threshold quanta by the photoelectron. The probability of population of high ATI levels due to this mechanism decreases rapidly, but it remains much higher, exponentially higher, than the probability of absorption of the same number of quanta due to direct ionization of the second electron. The return of the photoelectron to the parent atomic particle and inelastic scattering are possible for any ATI level excited due to the direct mechanism Ref. [5]. It makes the scattering mechanism productive.

There is the other possibility for the population of high ATI levels. Consider the following scenario. First the photoionization takes place. Then the scattering of the photoelectron results in the population of a high level in the ATI spectrum. After that the second collision takes place resulting in the double ionization. Analyzing the classical trajectory it is easy to show that the possibility for the second collision depends on the energy level in the ATI spectrum. If this level is below $f^2/2$ then the second collision can take place. Otherwise, the photoelectron leaves the atomic particle after the first collision and the considered scenario gives no contribution to the excitation of the second electron. Therefore this scenario works only in the restricted energy range. The given consideration shows that the inelastic collision

is possible for $k < f$. According to Eq.(2) it means that the ionization energy is restricted by $E^{(A^{++})} - E^{(A^{+})} \leq 2f^2$.

In order to demonstrate the validity of the presented results, consider the double ionization of an He atom. In the recent experiment Ref. [8] the 780 nm laser field with intensity from 0.15 to 5.0 PW/cm^2 was used. Even for the lowest intensity the yield of the doubly charged ions was very pronounced. For intensity 0.15 PW/cm^2 the field momentum is $f = 1.11$. Therefore the maximal wiggling energy, $f^2/2 = 17.0$ eV is well below the lowest excitation energy of He^+, which is 40.8 eV. Absorption of only 3 quanta above the single-electron ionization threshold changes the situation drastically. It gives the translational momentum $k = 0.642$. As a result the maximal energy of the photoelectron, $(k + f)^2/2 = 42.2$ eV, becomes higher than the excitation energy allowing the excitation of He^+. Similarly, absorption of 6 - 7 quanta above the atomic threshold makes the double ionization possible. Fig.1 shows the terms of (e+He^+) and (e_1+e_2+He^{++}) systems given by Eqs.(3),(4). Absorption of 7 quanta above the atomic threshold permits crossing of the term **b** of (e+He^+) with the terms **d**,**e** of (e_1+e_2+He^{++}). Thus the latter can be populated and double ionization takes place. The probability of absorbing a few quanta above the threshold is very high for the considered region of intensities of the laser field, see for example Ref. [7]. Notice that there is the damping factor which describes how small is the cross section of an inelastic collision in the laser field compared with the region of electron localization $\sigma_{\rm in}/R^2$. For estimation of the cross section one can use the value $\sigma_{\rm in} \approx 5 \times 10^{-18}$cm^2, which according to Refs. [22], [23]) describes the ionization as well as the excitation field-free cross sections. If we estimate the region of electron localization with the help of Eq.(6) then for the intensity 0.15 PW/cm^2 for 3 above-threshold quanta we find $R = 48a_0^2$, which gives $\sigma_{\rm in}/R^2 \approx 0.7 \times 10^{-4}$. In Ref. [8] the same ratio was estimated using the modification of the model Ref. [6]. This estimation gave the similar result 1.5×10^{-4}. In a recent work Ref. [9] there was made an attempt to consider quantitatively the role of the rescattering mechanism in the formation of the doubly charged ions of Helium. With this purpose the classical model was developed. The calculations based on this model give the probability of the He^{++} formation which is approximately by two orders of magnitude lower than the experimental value of Ref. [8].

Notice that the rescattering mechanism in any case gives a huge enhancement compared with the sequential ionization. There is still a discrepancy between the numerical and the

experimental results. It was interpreted in Ref. [9] as an indication that the rescattering is not important for the considered case, and it was suggested that the violation of the adiabatic conditions during the removal of the first electron can be responsible for the effect. There is a general physical argument against such a conclusion. In the initial wave function of Helium the separation of the energy levels greatly exceeds the frequency of the laser field. Therefore the adiabatic condition $\omega \ll \Delta E$ is well fulfilled. In this case the adiabatic theory predicts a very strong, exponential suppression of transitions from the ground state into excited states, see Ref. [20]. The situation drastically changes after the ionization of the first electron, when crossings of the terms become possible. At the moment of the crossing the adiabatic condition is clearly violated which results in the population of the higher terms and provides the possibility for the double-charged ion production. Thus the rescattering is to be considered as the preferred mechanism to explain the phenomena observed in Ref. [8]. This statement seems to agree with the results of calculations reported quite recently in Ref. [21].

There are several points for uncertainties in all the calculated or estimated values mentioned above. One of them is the poor knowledge of the inelastic cross sections in the laser field. The other one is the model nature of the developed approaches. The main assumption of this work, as well as of Ref. [5], is the neglect of the Coulomb field. From the physical point of view it is reasonable to assume that the Coulomb attraction can only but enhance the probability of two-electron processes.

After single electron ionization of the atom the term of the system $(e + A^+)$ in the laser field can undergo a crossing with some term of the system $(e_1 + e_2 + A^{++})$ (or $(e + A^{+*})$). As a result a transition from the first term to the second one is allowed and the second term is highly populated. This means that two-electron processes are very probable. ATI plays a very important role in this mechanism due to two reasons. First, it results in a rise of the maxima of the terms of the system of $(e + A^+)$ after absorption of only a few quanta above the atomic ionization threshold. Second, the ATI states above the threshold of doubly charged ion give the strong decrease of minima of the terms of the doubly charged ion.

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FIGURES

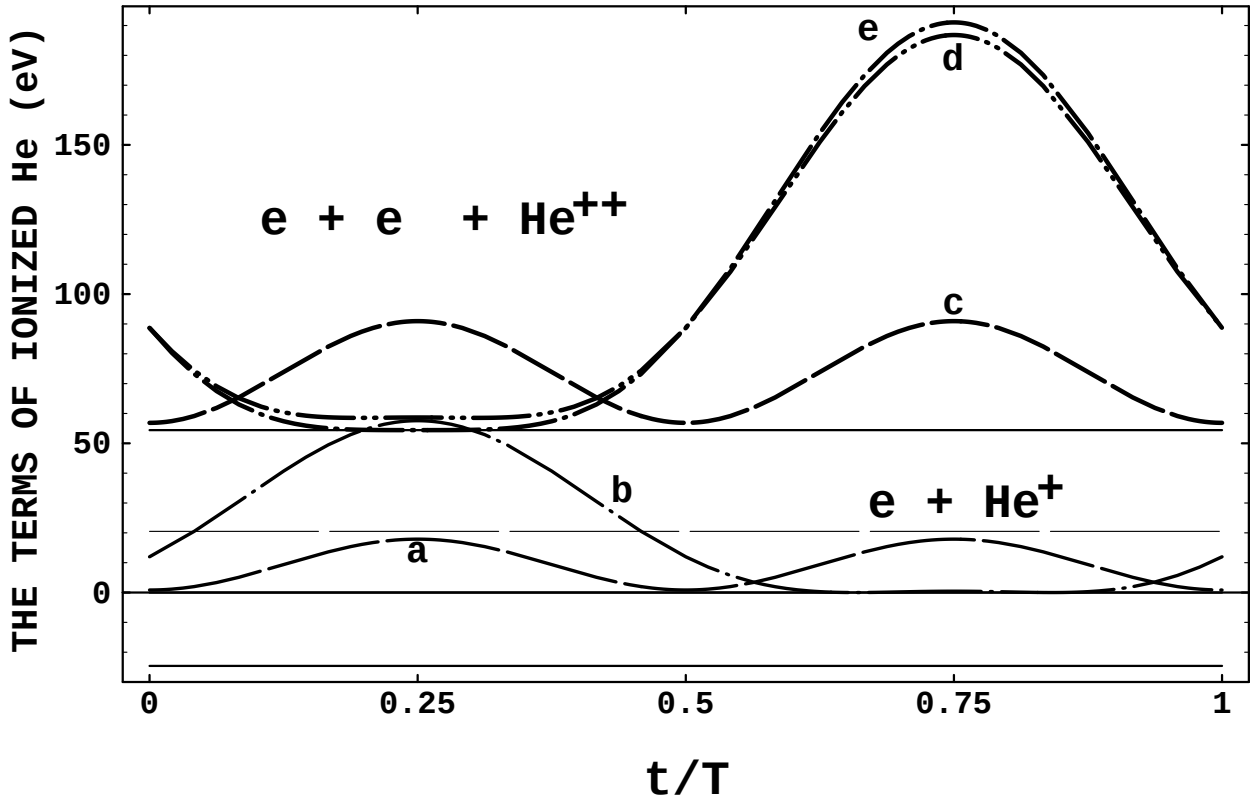

FIG. 1. Time evolution of the terms of the systems (e+He^+) and (e+e+He^{++}) given by Eqs.(3),(4). $\omega = 2\pi/T = 1.59$ eV corresponds to $\lambda = 0.780$ nm. The field strength $F = 0.0654$ $a.u$ corresponds to the lowest intensity 1.5×10^{14}W/cm^2 in experiment Ref.[8]. The energy levels of He, He^+ and He^{++} are shown by the horizontal solid lines. **a**.The "unfavorable" term of (e+He^+): the lowest level above the atomic threshold is occupied, the available translational momentum is perpendicular to the field. **b**.The "favorable" term of (e+He^+): 7 above-threshold quanta are absorbed and the translational momentum is parallel to the field. The horizontal dashed line is the position of this level in the ATI spectrum. **c**.The "unfavorable" term of (e+e+He^{++}): both electrons occupy the lowest above-threshold level, their translational momenta are perpendicular to the field. **d**,**e**. The "favorable" terms of (e+e+He^{++}): 20 quanta above the threshold of He^{++} are absorbed, the translational electron momenta are opposite to the field. **d** and **e** differ in the the distribution of the above-threshold energy E_1+E_2= 20 ω between the two electrons. **d**.$E_1 = E_2 = 10\omega$. **e**. $E_1 = 3\omega$, $E_2 = 17\omega$. Absorption of 7 quanta above the He threshold gives the crossing of the lower term **b** with the upper terms **d** and **e** resulting in double ionization.